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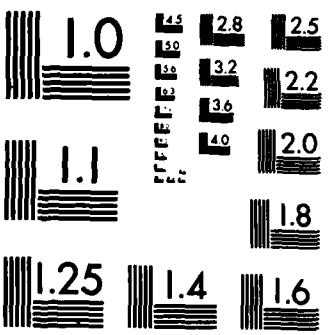

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STRUCTURE FORMATION IN ELECTRO-RHEOLOGICAL SUSPENSIONS IN AN ELECTRICAL FIELD I. QUALITATIVE CONSIDERATIONS

by

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STRUCTURE FORMATION IN ELECTRO-RHEOLOGICAL SUSPENSIONS IN AN ELECTRICAL FIELD I. QUALITATIVE CONSIDERATIONS

[STRUKTUROOBRAZOVANIE ELEKTROREOLOGICHESKIKH SUSPENZII V ELEKTRICHESKOM POLE I. KACHESTVENNOE RASSMOTRENIE]

by

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AUTHOR'S SUMMARY

Based on the formerly conducted studies of electro-rheological suspensions and model particles, the reasons for an electro-rheological effect are discussed. An attempt has been made to follow the connection between the formation of strong bridge structures of particles in an electric field determining the character of the mechanical behaviour of suspensions and the peculiarities of charge transport in an adsorption activator on the surface of silica gel particles.

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1 TEXT

The analysis of general regular movements of particles in suspensions[1-3], the mechanical behaviour in the shear flow of rods[4], of doublets[5], chains[6], aggregates[7], ellipsoids[8,9], and also the effect of an electrical field on the viscosity of a suspension of dielectric particles[10,11], has shown that, on the basis of information available, it is impossible to explain even the order of magnitude of the effective viscosity of electro-rheological suspensions upon application of an electrical field. Attempts to produce rheological models of flow in such suspensions[12] or to create a theoretical model of the flow of a suspension structurally formed in an electrical field[13] have so far afforded no useful results.

The visual observations and investigations on model particles which we have carried out have shown:-

(a) that, when an electrical field is applied the particles of an electro-rheological suspension oscillate across a gap and form aggregates and bridges[14];

(b) the movement of particles of silica in a non polar medium is entirely determined by the type and quantity of the polar compound, the activator, adsorbed on the surface of the particles;

(c) the bridges of particles adjacent to the electrodes have a definite stability, depending on the field strength and on the quantity of activator[15];

(d) the movement of particles is governed by the charge transfer across the gap;

(e) in an electrical field particles of silica-plus-activator behave in a manner similar to that of conducting substances but charge transfer in the adsorbed layer has an ambiguous character[16].

A change in the rate of deformation of the shear flow affects not only the size, but also the character of the change in the effective viscosity. In the majority of suspensions in the range of small shear rates (approximately $\dot{\gamma} \approx 150 - 200\ s^{-1}$), the value of η_{eff} falls very sharply; as $\dot{\gamma}$ increases further, the effective viscosity decreases slightly. It is natural to suppose that, for low shear rates, a factor which has a basic effect on the mechanical behaviour of a suspension is the structural formation of bridge type systems along the lines of force of the electrical field, *ie* across the flow. This is possible when the effects of both fields (the electrical and hydrodynamic) coincide. In the case of pressurised flow, the bridges are subjected mainly to bending stresses. In the presence of Couette flow, the movement of one of the electrodes produces only an expansion of the bridges, but this, in its turn, increases the shear stress in the electro-rheological suspension. For low shear rates in the suspension, a large number of bridges can form adjacent to the electrodes and the shear resistance is large. However, the bridge connected to the electrodes in time breaks and disintegrates. The particles released from these bonds begin to reorganise themselves into a new structured formation. With an increase in the shear rate, the life of each bridge, and with it the number of bridges existing simultaneously, decreases and the shear

deformation resistance of the suspension falls. Above a certain deformation rate, the shear stress hinders the formation of bridges, particularly in suspensions of low concentration. An increase in the effective viscosity in an electric field (which is usually small), in the range of moderate rates of shear deformation is determined by the interaction of polarised particles with the external field and with each other. Apparently, the greatest effect on the effective viscosity in this case must be exerted by the stability and number of bridges and also by the polarised interaction of the particles involved in the structure formation.

In our tests with silica gel granules activated by moisture and by diethylamine, it was shown[15] that the force required to tear a particle from an interelectrode bridge, like the effective viscosity of the suspension, bears an almost parabolic relationship to the field strength and an almost linear relationship to the size of the current flowing across the bridge. A rise in the activator content (and with this also a rise in the conductivity of the particles) at first increases the separating force and then progressively decreases it. Thus the formation of a bridge is accompanied by a significant increase in conductivity; this implies that the electro-rheological effect is encountered only in the presence of a finite conductivity of the suspension. A lowering of the stability of the bridges (and a lessening of the increase in the effective viscosity observed) following the attainment of a conductivity greater than a certain optimum value can be regarded as evidence of the fact that the basic contribution to the stability of bridges is made by contact phenomena at the particle-particle and particle-electrode boundaries. In the contact zone, there must exist a definite conductivity and at the same time a certain potential difference giving rise to additional compression of the particles.

An analysis of experimental data available makes it possible to put forward the proposition that, in the adsorbed layer of activator on the surface of the silica particles, there must exist at least two types of charge carriers with different mobilities. One of these ensures a current across the bridge for small amounts of activator; the charge transfer signals of platinum particles are identical in nature to the discrete pulses of the charge transfer signals of activated silica gel. It also ensures the independence of the form and geometry of the charge transfer signals of silica gel on the sign of the electrode, (*ie* the fact that the work of transferring the charge from a metal electrode to a particle of silica is the same as that of transferring the charge from a particle of silica to the electrode). Such a charge carrier can be an electron. The second charge carrier, less mobile, can be considered as ionic. Provided that the substance of the activator is capable of forming hydrogen bonds, it is also possible to postulate proton conductivity.

Ionic charge transfer governs:

(a) the complex multipulse form of the charge transfer signal of a silica gel particle. Actually, when an oscillating particle touches the electrode, in the first moment a certain number of electrons transfer to the latter with the surface of the particle turned towards the electrode. The potentials of

this surface and the electrode equalise. The remaining electrons associated with less mobile ions are not discharged. The particles withdraw to a certain distance from the electrode and the charges are redistributed over the surface of the particle. On the face turned towards the electrode there appears a charge of opposite sign and the particles are again attracted to the electrode;

(b) the compression of particles in the bridge, which can be explained by the presence of a certain difference in potentials between particles in their contact zones;

(c) the qualitatively similar change in many properties upon heating, in particular the increase in the effective viscosity, in conductivity, in relative dielectric constant and in the tangent of the angle of dielectric loss of a suspension of such particles with an increase in temperature (due to the increase in mobility of the ions).

The presence of charge carriers and of their specific features mentioned above are determined by the construction of an absorbed layer of activator - water, amine, alcohol, acid - on the surface of the silica. The majority of data relates to the structure of an adsorbed layer of water. Known investigations into heats of adsorption, infra red spectra, dielectric properties, thermal conductivity of hydrated disperse (and layer) systems are described[17,18]. From these, it follows that the first portions of water adsorbed on the most active centres of the surface of silicas have a strictly orientated structure with little conductivity or polarisation but with a high thermal adsorption. In an electro-rheological suspension, particles with a thin closed adsorbed layer are poorly electro-conducting, while, at the same time, they show an ability to increase significantly the effective viscosity in an electrical field. The charge transfer signals of such particles consist of several sharply defined pulses; the value of the current across an individual bridge does not depend on time[16,19]. As was shown[20], on the surface of certain disperse materials in aqueous medium, a layer of unipolar orientated molecules of water is formed, resulting in the particles acquiring a stable dipole moment. According to published data[21], the first molecules of water when adsorbed on silica form coordination bonds with hydrated atoms of silicon. In this case, there is found to be a displacement of the centre of negative charges of the silanol group - water molecule system towards the silicon atom.

The totality of the findings described above has still not been generalised in the form of a structural model of the adsorbed layer of water. As a hypothesis, it is possible to consider the polar activator, particularly water, in the layer directly adjoining the surface of the silica as a capacitor of molecular dimensions with an inner (surface) plate having electron conductivity and an outer plate having ionic conductivity. It is difficult to say anything definite concerning the thickness of this layer at the present time. Apparently it is a bi- or tri-molecular layer formed as the result of the merging of surface clusters. Due to the energetic non-uniformity of the surface of dehydrated silica, adsorption of water occurs initially on the most active centres in the form of clusters which then coalesce into a dense layer[17]. This layer is strictly

orientated; all impurities are expelled from it. Further filling-in occurs as the result of adsorption of subsequent quantities of water on the first layer. The effect of the surface of the silica decreases in this case. The structure of subsequent layers is therefore less ordered than that of the first layers and the adsorbed molecules have a greater freedom of movement. Because of this, there is a large increase in the dielectric constant and particularly in the tangent of the angle of dielectric loss in the low frequency range, in the conductivity[22], and in the effective thermal conductivity[23] of suspensions under stress with a further increase in the thickness of the adsorbed layer of water on the particles. Tests on the oscillation of individual particles have shown that the properties of the adsorbed layer of water change smoothly as it becomes further removed from the surface of the particle, while, if diethylamine is used, the change is abrupt. In the model of a flat capacitor, the outer plate changes its properties depending on the quantity of adsorbed compound and on the temperature. Thus, at room temperature for a bridge of suspended particles with a closely adsorbed layer of minimum thickness, the value of the current flowing does not depend on time. Upon heating, this same bridge shows a decrease of current with time; the thermal movement so to speak 'loosens' the outer part of the strictly orientated layer[19].

The application of a uniform electrical field to a suspension of activated particles of silica in a non-polar medium produces a redistribution of charges on the surface of the particles. The field strength in the volume of the particles decreases, while, in the gaps between them it increases in comparison with the initial strength. Pondermotive forces displace the polarised particles in the region of high field strength either towards the electrodes (oscillation commences) or one towards one another (aggregates then bridges are formed). The formation of bridges occurs when charge transfer based on conductivity is energetically more favourable than charge transfer based on oscillation or electro-convection. In a bridge of particles having the optimum amount of activator, under the effect of the field, compression of particles occurs until the outer plates of the capacitor are joined (an electron flow, the value of which is independent of time, passes through a chain of joined plates). The positive ions displaced within the limits of each particle, towards the negative electrode cannot be discharged at the metal electrode. Therefore, remaining in the activator layer, they create a potential difference in the particles, thereby increasing the compression of the bridge (and this, in its turn, increases the stability of bridges). Thus the hypothesis concerning the dual nature of the conductivity of the adsorbed layer makes it possible to draw up a theoretical interpretation of known facts which correctly agrees with measurements and observations; also, it is possible to relate it to the increase in effective viscosity and stability of bridges and to the current flowing through the system and thereby establish the proportionality of the above parameters to the square of the field intensity, which usually characterises polarised interactions of particles.

For an excess of activator (ϕ) above the optimum quantity, the structure of the outer part of the adsorbed layer is more brittle and mobile than that of the surface layer. The inter-relationship of the two activator layers should be considered; all processes which occur in one layer have an effect on the charge transfer parameters in

both layers. An excess of activator above the optimum quantity gives rise to the diffusion and high conductivity usual for water. Moreover, in a strong electrical field, a displacement of activator molecules is possible into regions of elevated field intensity, more precisely into zones of contact between particles themselves or between particles and electrodes. This process is not instantaneous and produces a gradual increase in the parasitic conductance in contact zones accompanied by a fall in the cohesive force between particles in bridges. Charge transfer signals of silica particles at electrodes consist of a few weak pulses for low activator concentrations; when ϕ_{opt} is obtained, the pulses become sharply defined, their number attaining 4 or even 5. In the region $\phi > \phi_{opt}$, an increase in the intervals between pulses is observed and they degenerate until there remains but a single charge transfer pulse of the very moist silica. The special features of charge transfer which have been examined and which are governed by the existence of a molecular capacitor in the adsorbed layer of water, amine, alcohol, or acid, on the surface of silica, are detected only in strong fields and over a limited range of concentration of the substance adsorbed. The conductivity in a layer of activator weakly bound to the surface is manifest even in low fields. Conductivity is naturally possible in adsorbed layers of activator-type compounds on surfaces not possessing prominent charges, for example on polytetrafluoroethylene. In the adsorbed layer of polar activator on silica, the effect of the surfaces gradually weakens. Therefore, in a strong field, with suspensions having an increasing quantity of activator, one form of conductivity (electron) gradually gives way to another (ionic) but no sharp transition is detected.

It is possible from what has been said to conclude that two types of inter-related charge carriers, present in the volume of the particles or introduced with other compounds - activators - can be attributed to particles of electro-rheological suspensions; this is in contrast to the case of other polarising chains of particles formed in an electric field. One of the charge carriers experiences little resistance in zones of contact, *ie* ensures conductivity through the bridges; the second charge carrier, however, is associated with high contact resistance. There is a definite analogy between the behaviour of particles of electro-rheological suspension in the interelectrode gap and of a semi conductor - metal system with the metal under stress. In recent times, the production of compressive forces known as the Johnson-Rabek effect[24], has been observed.

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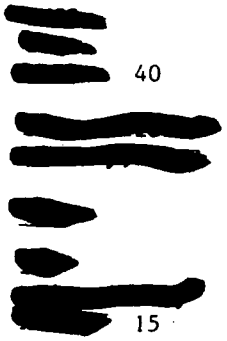
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